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SECURITY CLASSIFICATION OF THIS PAGE

(2)

PORT DOCUMENTATION PAGE

DTIC FILE COPY

AD-A203 247

1b. RESTRICTIVE MARKINGS

3. DISTRIBUTION/AVAILABILITY OF REPORT
Approved for public release; distribution unlimited.

4. PERFORMING ORGANIZATION REPORT NUMBER(S)
SR88-24

5. MONITORING ORGANIZATION REPORT NUMBER(S)

6a. NAME OF PERFORMING ORGANIZATION
Armed Forces Radiobiology Research Institute

6b. OFFICE SYMBOL (If applicable)
AFRRI

7a. NAME OF MONITORING ORGANIZATION

6c. ADDRESS (City, State and ZIP Code)
Defense Nuclear Agency
Bethesda, Maryland 20814-5145

7b. ADDRESS (City, State and ZIP Code)

8a. NAME OF FUNDING/SPONSORING ORGANIZATION
Defense Nuclear Agency

8b. OFFICE SYMBOL (If applicable)
DNA

9. PROCUREMENT INSTRUMENT IDENTIFICATION NUMBER

8c. ADDRESS (City, State and ZIP Code)
Washington, DC 20305

10. SOURCE OF FUNDING NOS.

PROGRAM ELEMENT NO.	PROJECT NO.	TASK NO.	WORK UNIT NO.
NWED QAXM			

11. TITLE (Include Security Classification)
(see cover)

12. PERSONAL AUTHOR(S)
Miller, J. H., Wilson, W. E., Swenberg, C. E., Myers, L. S. Jr., Charlton, D

13a. TYPE OF REPORT	13b. TIME COVERED	14. DATE OF REPORT (Yr., Mo., Day)	15. PAGE COUNT
Reprint	FROM ____ TO ____	1988 November	5

16. SUPPLEMENTARY NOTATION

17. COSATI CODES		
FIELD	GROUP	SUB. GR.

18. SUBJECT TERMS (Continue on reverse if necessary and identify by block number)

19. ABSTRACT (Continue on reverse if necessary and identify by block number)

DTIC ELECTE DEC 23 1988 S D E

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A-1 | 20

20. DISTRIBUTION/AVAILABILITY OF ABSTRACT
UNCLASSIFIED/UNLIMITED ☐ SAME AS RPT. ☐ OTIC USERS ☐

21. ABSTRACT SECURITY CLASSIFICATION

22a. NAME OF RESPONSIBLE INDIVIDUAL	22b. TELEPHONE NUMBER (Include Area Code)	22c. OFFICE SYMBOL
M. E. Greenville	(202) 295-3536	ISDP

DD FORM 1473, 83 APR EDITION OF 1 JAN 73 IS OBSOLETE.

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SECURITY CLASSIFICATION OF THIS PAGE

Radiat. Phys. Chem. Vol. 32, No. 3, pp. 349–353, 1988
Int. J. Radiat. Appl. Instrum. Part C

0146-5724/88 $3.00 + 0.00

MODELING RADICAL YIELDS IN ORIENTED DNA EXPOSED TO HIGH-LET RADIATION

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(*Received* 16 *June* 1987)

Abstract—Monte Carlo simulation of energy absorption in oriented fibers of DNA is used to model the dependence of free radical yields on the orientation of the fibers relative to a flux of ionizing radiation. We assume a large asymmetry in the thermal conductivity of the fibers that permits rapid transport of vibrational energy along a DNA molecule, but not between different molecules. Based on this assumption, our model predicts that thymine radical anions have a significantly greater probability of undergoing a secondary protonation reaction if they are produced by a flux of high-energy protons that is incident parallel to the helical axis of DNA than if they are generated by a flux that is incident perpendicular to the DNA molecules. These results are in qualitative agreement with experimental data on the yield of 5,6-dihydrothymin-5-yl radicals when samples of oriented DNA were exposed at 77 K to neutrons.

Keywords: High linear energy transfer, LET (Linear Energy Transfer), High Energy Protons, Prints.

INTRODUCTION

Understanding the mechanisms by which high-energy charged particles produce reactive chemical species in biologically significant materials is important for assessing the health effects of low-level exposure to ionizing radiation. This is particularly true for radiation with high linear-energy-transfer (LET) where correlations between track structure and target structure can have a profound effect on the nature and distribution of damage. The role of nonhomogeneous processes in radiation chemistry was recently reviewed by Magee and Chatterjee.[1] Our research in this area[2] has focused on the use of detailed Monte Carlo simulation of the spatial distribution of energy absorbed from charged particles[3] as a basis for understanding the subsequent evolution of chemically active species. In this paper we report the use of this approach to investigate production of free radicals in oriented DNA exposed to a flux of high-energy protons.

Samples of oriented DNA fibers prepared by the wet spinning technique have proven to be a valuable experimental tool for investigating the production of free radicals in DNA by both u.v. light and ionizing radiation.[4–7] The recognition that spatially correlated electron gain and loss centers may be precursors of DNA double strand breaks[8] has led to renewed interest in this experimental system for investigating radiation-induced DNA damage. Recently, Arroyo *et al.*[9] used this type of sample in conjunction with electron spin resonance (ESR) to investigate the production of radicals at 77 K by exposure to neutrons. When samples were irradiated by a flux of neutrons incident perpendicular to the helical axis of DNA, ESR spectra showed that thymine anion (T^-) and guanine cation were the principal radical species present at 77 K. However, when the neutron flux was approximately parallel to the orientation of DNA molecules, the total yield of radicals was reduced and a new ESR signal characteristic of the 5,6-dihydrothymin-5-yl ($TH^\cdot$) radical predominated.

The most obvious difference in the patterns of energy absorption in the two irradiation geometries is that charged particles that traverse the sample nearly parallel to the helical axis of DNA have a greater probability of multiple energy transfer to the same molecule than do particles incident on the sample perpendicular to the fiber orientation. This difference in the spatial distribution of energy absorption should have no effect on the production of free radicals if energy and/or charge move between DNA molecules in a fiber as freely as they are transported along a single DNA chain. Hence the unusual orientation dependence of radical yields in this system may have important implications for intramolecular energy and charge transfer in DNA.

Arroyo *et al.*[9] speculated that the reduction of radical yields in the parallel irradiation geometry was due to enhanced recombination of precursor ion pairs. This explanation seems plausible in light of recent measurements of conductivity by van Lith *et al.*[10] which suggest that the high mobility of electrons in the ice-like water layer around a biopolymer could enhance intramolecular charge transfer. Arroyo *et al.* attributed the appearance of $TH^\cdot$ radicals in the parallel irradiation geometry to intramolecular transfer of triplet excitation produced in charge recombination. The long lifetime of triplet excitons at 77 K[11] certainly favors this mechanism. However, due to the variation in triplet transfer rates,[12] triplet

excitons tend to be restricted to the neighborhood of thymine that is bounded by the nearest guanine or cytosine on each side. Hence, in DNAs with roughly equal G-C and A-T content, the range of triplet migration is not expected to be large. Ion quenching experiments[13] suggest a range of 5–30 base pairs. The short lifetime of singlet excitons[14] is probably the major factor that limits the range of their migration.

In this paper we present a model for the orientation dependence of $TH^{\cdot}$ yields that is based on an assumed asymmetry in the transport of vibrational excitation in the oriented DNA samples. Asymmetry of phonon scattering rates in oriented DNA have been observed using Raman spectroscopy;[15,16] however, these low-frequency modes should not be very effective in activating proton transfer to $T^{\cdot-}$ if the activation energy is 0.2–0.7 eV as suggested by the work of Gräslund *et al.*[5] Regardless of the mechanism, our model is macroscopic in that asymmetric energy transfer is approximated by the solution of thermal diffusion equations with a coefficient of diffusion along DNA that is 1000 times greater than the coefficient for diffusion perpendicular to the helical axis. This model is developed in the next section of the paper and our numerical results for the production of $TH^{\cdot}$ radicals by proton irradiation at 0.3 and 1.0 MeV are discussed in the third section. Conclusions and the potential for improved theory and experiment are discussed in the final section.

THEORY

As suggested by Gräslund *et al.*,[5] we assume that the major source of $TH^{\cdot}$ radicals is the reaction

$$T^{\cdot-} + XH^{+} \rightarrow TH^{\cdot} + X, \tag{1}$$

where the proton donor, XH^{+}, is probably water of hydration. In an excess of proton donor, this reaction will be pseudo first-order and the yield of radiation-induced $TH^{\cdot}$ radicals per DNA molecule is given by the equation

$$Y_{TH} = \int_{-x}^{+x} \pi b^2 \, dx C_{T^{\cdot-}}(x, 0) P_{TH^{\cdot}}(x), \tag{2}$$

where

$$P_{TH^{\cdot}}(x) = \int_0^{t_{max}} k(x, t) \exp\left[- \int_0^t k(x, t') \, dt' \right] dt \tag{3}$$

is the probability that a thymine anion formed at position x on an oriented DNA molecule, which we approximate as a long cylinder of radius $b = 10$ Å, will be converted to $TH^{\cdot}$ by protonation. The initial concentration of $T^{\cdot-}$, the rate of protonation, and the duration of the radiation-induced excess temperature are denoted by $C_{T^{\cdot-}}(x, 0)$, $k(x, t)$, and t_{max}, respectively.

Modeling the production of $T^{\cdot-}$, which probably involves the decay of highly excited states through autoionization channels followed by capture of low-energy secondary electrons in traps with high electron affinity, is beyond the scope of this paper. Rather we simply assume that the probability to form $T^{\cdot-}$ in the ith energy deposition event is proportional to the amount of energy ϵ_i absorbed locally in that event. Hence, the initial concentration of $T^{\cdot-}$ is approximated by the equation

$$C_{T^{\cdot-}}(x, 0) = g_{T^{\cdot-}} \sum_i \epsilon_i \frac{\delta(x - x_i)}{\pi b^2}, \tag{4}$$

where $g_{T^{\cdot-}}$ is the average yield of $T^{\cdot-}$ per unit of absorbed energy and $\delta(x)$ is the Dirac delta function. Using this approximation in equation (3) we can perform the space integration and obtain the result.

$$\frac{g_{TH^{\cdot}}}{g_{T^{\cdot-}}} = \frac{\sum_i \epsilon_i P_{TH^{\cdot}}(x_i)}{\sum_i \epsilon_i}, \tag{5}$$

for the yield of $TH^{\cdot}$ relative to the yield of its precursor $T^{\cdot-}$. This result is just a weighted average of the probability to convert $T^{\cdot-}$ to $TH^{\cdot}$ over all the positions of energy deposition in a DNA molecule from charged particles traversing the sample at a specified angle relative to the molecular orientation. The weighting factors express the relative probability that $T^{\cdot-}$ will be produced by a given energy absorption event.

The rate of proton transfer used in equation (3) to calculate $P_{TH^{\cdot}}$ depends upon the transient local temperature $T(x, t)$ through the relation

$$k(x, t) = A \exp[-Q/T(x, t)], \tag{6}$$

where A is a constant of the order of vibration rates (10^{13}/sec) and Q is the activation energy. If we completely neglect intermolecular transfer of vibrational energy and make the additional assumption that the heat capacity and thermal conductivity are independent of temperature, then $T(x, t)$ can be obtained by solution of the one-dimensional thermal diffusion equation for a cluster of point sources.[13,14] This result can be written in the form

$$T(x, t) = T_s + \sum_k T_k(x, t), \tag{7}$$

where T_s is the ambient sample temperature,

$$T_k(x, t) = T_{k0}(1 + t/\tau_{\uparrow\uparrow})^{-1/2} \exp\left[\frac{-(x - x_k)^2}{2\Delta^2(1 + t/\tau_{\uparrow\uparrow})} \right], \tag{8}$$

with

$$\tau_{\uparrow\uparrow} = \frac{\Delta^2}{2D_{\uparrow\uparrow}} \tag{9}$$

being the parallel diffusion relaxation time and

$$T_{k0} = \frac{\epsilon_k}{\sqrt{2\pi^3 b^2 \Delta \rho C}} \tag{10}$$

being the initial excess temperature at x_k due to absorption of energy ϵ_k. The sample density, specific heat and coefficient of thermal diffusion parallel to

the DNA fibers are denoted by ρ, C and $D_{\uparrow\uparrow}$, respectively. Ultimately C and $D_{\uparrow\uparrow}$ must be considered as adjustable parameters since we are using macroscopic material parameters to describe a microscopic process. In addition, the effective specific heat C contains a factor for the conversion of energy from electronic to vibrational excitation. As a starting point for numerical calculations, we use the specific heat and thermal diffusion coefficient of water [4×10^7 erg/(g*deg) and 10^{-3} cm^2/s, respectively]. The delocalization of vibrational excitation by subpicosecond processes is included in our model through the parameter Δ, which is the initial width of the Gaussian distribution that approximates the excess temperature induced by individual energy deposition events. We expect Δ to be of the order of the base stacking distance which is 3.4 Å in the B conformation.

The formulation of the model thus far completely neglects transport of vibrational energy perpendicular to the helical axis of the DNA molecule.This is generally adequate for small isolated clusters of excitation where the radiation-induced excess temperatures decays on a nanosecond or smaller time scale. However, when many interactions occur over an appreciable length of DNA the time dependence of the excess temperature may be affected by thermal-diffusion perpendicular to the fiber direction. Since we are assuming that $D_{\uparrow\uparrow} \gg D_{\perp}$, it is reasonable to approximate this leakage by considering the loss of thermal energy from a long cylinder of uniform initial temperature. Including only the first term in the series expansion that is the solution of this well-known heat flow problem, we obtain an approximation for the transient local temperature that is given by

$$T(x, t) = T_s + \exp\left(-\frac{t}{\tau_{\perp}}\right) \sum_k T_k(x, t), \qquad (11)$$

where

$$\tau_{\perp} = \frac{b^2}{5.8 D_{\perp}} \qquad (12)$$

is the perpendicular thermal diffusion relaxation time. If the asymmetry in thermal diffusion is large (we assume $D_{\perp}/D_{\uparrow\uparrow} = 10^{-3}$) and if the initial width parameter does not greatly exceed the radius of the molecule, then $\tau_{\perp} \gg \tau_{\uparrow\uparrow}$ and the leakage factor in equation (11), $\exp[-t/\tau_{\perp}]$, serves only to limit the duration of the excess temperature when many excitations occur in the same molecule. Hence it mainly effects the yields calculated in the parallel irradiation geometry.

RESULTS

Application of the formalism presented in the previous section requires information about the spatial patterns of energy absorption in a DNA molecule of specified orientation relative to the flux of ionizing radiation. In our model this information consists of the positions of energy deposition events x_k and the amount of energy ϵ_k deposited locally in each event. Energy transported away from x_k by high-energy secondary electrons is not included in ϵ_k, since the detailed structure of these secondary tracks is included in our Monte Carlo simulation of the slowing down of the primary ion. To approximate the spatial distribution of energy deposited in a DNA molecule, we superimpose a long right-circular-cylinder of radius $b = 10$ Å on a computer-simulated segment of the track of a high energy proton slowing down in water. The computer simulation is carried out by a Monte Carlo code developed by Wilson and Paretzke.[3] The superposition of cylinders and track segments is random in all aspects except the angle between the axis of the cylinder and the velocity of the proton. The collection of energy transfer points in this superposition of target and track we will refer to as a "hit".

Once an energy deposition event within a hit has been randomly selected to be the site of formation of $T^{\cdot-}$, the transient temperature at that position is calculated for equation (11). Typical results for hits that involve small and large amounts of energy deposited in DNA are shown in Fig. 1. If $D_{\perp}/D_{\uparrow\uparrow} \leq 0.001$ then the leakage term in equation (11) does not significantly effect the transient temperature at times less than about 0.1 ns. However, transport of thermal energy perpendicular to the DNA fibers does influence the excess temperature associated with large hits at longer times and substantially reduces the probability that protonation will occur. Unless otherwise noted, all of the results presented in this paper are for $D_{\perp}/D_{\uparrow\uparrow} = 0.001$.

Assuming an initial delocalization of vibrational excitation equal to 3.4 Å, Fig. 2 shows the dependence of the yield of TH$^{\cdot}$ per $T^{\cdot-}$ precursor on the activation energy for the proton transfer reaction. In these calculations we have averaged over the distribution of hits that occur under a given irradiation

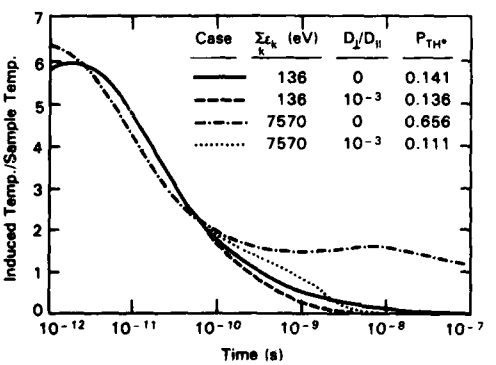

Fig. 1. Transient radiation-induced temperature changes for two typical hits with different spatial and energy characteristics. For times <0.1 ns temperature profiles are essentially the same for the two levels of asymmetry in thermal conductivity.

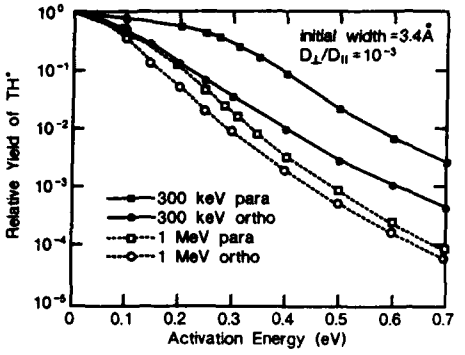

Fig. 2. Yield of TH• per T•- as a function of the activation energy for proton transfer.

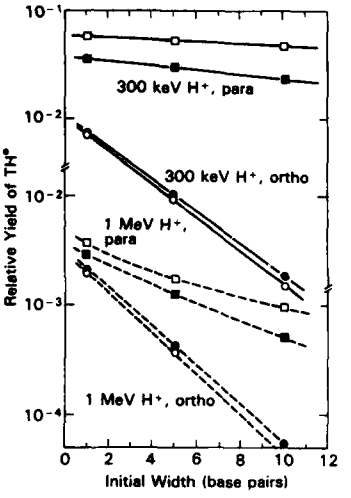

Fig. 3. Yields of TH• per T•- averaged over the distribution of activation energies given in Ref. 5 as a function of the initial width parameter of the model expressed in units of the base-pair separation of DNA in the B conformation (3.4 Å). Solid symbols show the effect of a $\pm 10°$ uncertainty in the orientation of DNA molecules relative to the proton flux.

condition. Typically, a sample containing on the order of 10^5 hits was examined. To obtain results for comparison with experiment we must further average the yield of TH• over a distribution of activation energies for the protonation reaction. If we use the distribution proposed by Gräslund *et al.*,[5] we obtain the results shown in Fig. 3. In this figure the ratio $g_{TH\cdot}/g_{T:}$ is plotted as a function of the initial width parameter in units of the base-pair separation, 3.4 Å.

Generally, protonation of T•- decreases exponentially with increasing initial delocalization of the vibrational excitation of the medium in an individual energy deposition event. This is expected from equation (10) which shows that the initial excess temperature associated with any energy deposition event is inversely proportional to the initial width Δ. This parameter also enters the calculation through equation (8) where the effect of a larger Δ is to increase the duration of the excess temperature, which favors protonation. Apparently the former effect predominates. In Fig. 3 we see that protonation of T•- in the parallel irradiation geometry is considerably less sensitive to the value of Δ than it is in the perpendicular case. In the parallel case a hit is made up of many more energy deposition events than in the perpendicular case. This makes the effectiveness of a hit for inducing protonation of T•- less dependent on the excess temperature associated with individual energy deposition events.

The effects on the yield of TH• of a $\pm 10°$ uncertainty in the orientation of the DNA fibers relative to the proton flux is also shown in Fig. 3. Results for the parallel orientation are more sensitive to this type of uncertainty than are those obtained for protons incident nearly perpendicular to the DNA fibers. This is a consequence of the fact that the large TH• yields in the parallel irradiation geometry result from the relatively few hits where a DNA molecule is close to the axis of a proton track for an appreciable distance. This aspect of our model is further illustrated by Table 1 which gives the average energy deposited in a DNA molecule by a hit and the root-mean-square separation of T•- from other energy deposition events in a hit. The latter quantity is a measure of the distance over which thermal energy diffuses in the process of activating proton transfer to T•-. Note that this energy transfer distance is essentially equal to the diameter of the target in the perpendicular case and is insensitive to a $\pm 10°$ uncertainty in this orientation. However, this level of uncertainty in orientation of DNA relative to a proton flux that is parallel to the fibers causes a large reduction in the average energy transfer distance and a correspondingly large reduction in the protonation of T•-.

Table 1. Characteristics of the interaction of oriented DNA with 1 MeV protons

Orientation[a]	Energy deposited (eV)	Transfer distance (Å)[b]
0	293	2240
0 ± 10	150	474
90	62	21
90 ± 10	62	22

[a] Angle in degrees between helical axis of DNA and proton velocity.
[b] Root-mean-square separation between sites of T•- and other excitations.

The characteristic of our model that is most readily testable is its dependence on the LET of the radiation. This aspect of our results is better illustrated by Fig. 4 where the ratio of yields of $TH^{\cdot}$ per $T^{\cdot-}$ precursor calculated with the same model parameters in the parallel and perpendicular case, is plotted as a function of the initial width parameter in units of the base-pair separation. This ratio is an indicator of the enhancement of the protonation reaction in samples exposed to a flux of high-LET radiation parallel to the orientation of the DNA fibers. In this model, the degree to which protonation of $T^{\cdot-}$ is enhanced in the parallel irradiation geometry is strongly dependent upon the initial spatial delocalization of vibrational excitation in the DNA. As was discussed above, this results from the fact that protonation of $T^{\cdot-}$ in the parallel case is less dependent on high local excess temperature. However, the increase in the enhancement factor with increasing LET is essentially independent of this initial width parameter. Hence experimental results at one proton energy can be used to determine Δ for comparison between theory and experimental at other proton energies. This approach to experimentally testing the model assumes that Δ is independent of LET.

CONCLUSIONS

Our model calculations show that intramolecular energy transfer following radiation exposure should enhance protonation of $T^{\cdot-}$ and that this is a reasonable explanation for the orientation dependence of $TH^{\cdot}$ yields that is observed when a flux of high-LET radiation is absorbed in samples of oriented DNA at 77 K.[9] The amount of enhancement predicted by the model is sensitive to the uncertainty in orientation and to the stopping power of the incident radiation. Both of these results point out the need to repeat the experiments of Arroyo *et al.*[9] with proton irradiation from an accelerator rather than a neutron source. This will reduce the uncertainty in orientation of the proton flux relative to the DNA fiber direction and allow the enhancement of $TH^{\cdot}$ radicals in the parallel irradiation geometry to be measured as a function of the LET of the incident radiation. Experiments of this type are in progress. Improvements in the model are being focused in two areas: (1) a more microscopic description of $TH^{\cdot}$ production with parameters that are more directly related to molecular properties of the system; and (2) a more complete theory that includes recombination which will allow us to predict both the enhancement of $TH^{\cdot}$ and the reduction of total radical yields in the parallel irradiation geometry. By this combined experimental and theoretical effort we expect that oriented DNA will be an even more useful tool for investigating the interplay of target structure and track structure in determining the production of reactive species in biological systems exposed to high-LET radiation.

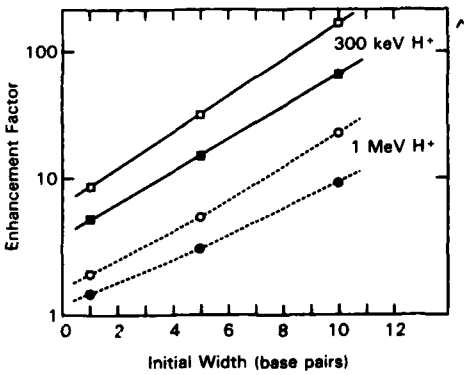

Fig. 4. Enhancement of protonation of $T^{\cdot-}$ formed by irradiation with a proton flux that is parallel to the orientation of DNA relative to the yield of this secondary reaction in the perpendicular irradiation geometry. Solid symbols show the effect of a $\pm 10°$ uncertainty in orientation of DNA molecules relative to the proton flux.

Acknowledgement—This work was supported by the Office of Health and Environmental Research (OHER) U.S. Department of Energy under Contract DE-AC06-76RLO 1830.

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